Supplementary Information for:

Combined effects of coagulation and ozonation treatment on landfill leachate DOM biodegradability

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Text S1 Optimization of coagulant conditions

Effect of dosage of coagulant

Figure S1 illustrates the COD degradation and turbidity changes observed for four coagulants across different dosage levels. During the coagulation process, the COD concentration in the supernatant initially decreases with increasing dosage but begins to stabilize or fluctuate within a specific dosage range. When PAC is employed as a coagulant, the COD concentration in the supernatant decreases gradually within the dosage range of 0.5–3 g/L. At a dosage of 4 g/L, the COD concentration decreases sharply, and the COD/PAC value, representing the COD removal per unit PAC dosage, reaches its maximum.

For PAFC, the COD concentration decreases steadily with increasing dosage and levels off at approximately 4 g/L. The COD/PAFC value initially rises and then declines, peaking at 4 g/L. When PFS is used as a coagulant, the COD concentration in the supernatant decreases consistently across the dosage range of 0.5–4 g/L, reaching its lowest value at 8 g/L. The COD/PFS value peaks at this dosage, indicating optimal removal efficiency.

Visual and physical characteristics of the treated leachates varied by coagulant. The PSAF-treated leachate appeared light green, with loose flocs that remained suspended near the water surface. PAC treatment resulted in a similar light green leachate, but the generated gray-green flocs were smaller and more difficult to settle. The PFS diluent exhibited a reddish-brown color, and upon addition to the leachate, the supernatant turned dark green with high chromaticity. The flocs generated during PFS treatment were the largest and most easily precipitated. PAFC-treated leachate appeared light reddish-brown with visible

coloration and loose flocs.

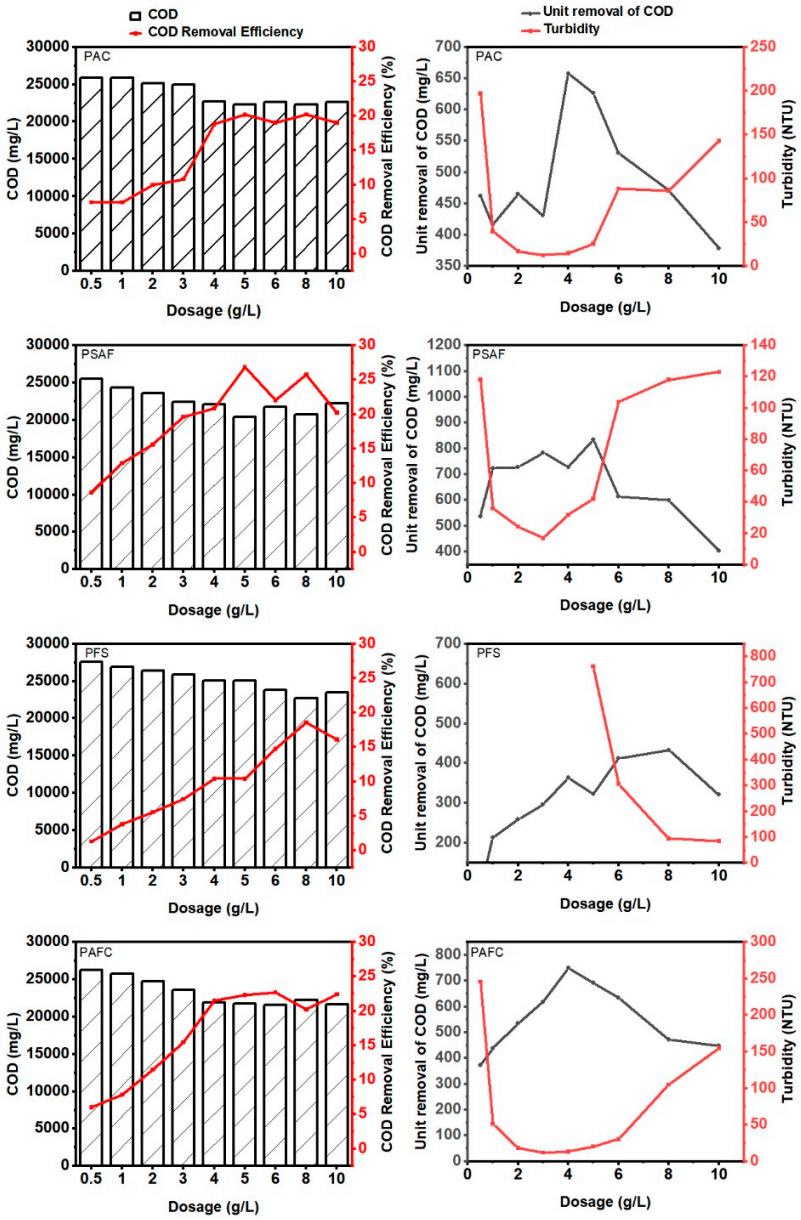


Figure S1. Effect of dosage of four coagulants on COD and turbidity in leachate (a-b PAC; c-d PAFC; e-f PFS; g-h PSAF)

Influence of dosage of flocculant

Flocculants are chemical agents that complement coagulants in water treatment processes, enhancing the aggregation and flocculation of colloidal particles. In this study, polyacrylamide (PAM) was primarily used as a flocculant, prepared as a 1 g/L aqueous solution prior to application. Figure S2 illustrates that increasing the PAM dosage initially led to a gradual increase in the COD removal rate for leachates treated with PAC, PAFC, and PSAF, eventually reaching a plateau. Concurrently, the turbidity showed a marked reduction.

As a cationic organic polymer flocculant, PAM neutralizes the negative charge on colloidal particle surfaces, inducing destabilization. Its polymer chains bridge suspended particles, forming larger flocs. Additionally, PAM adsorbs onto colloidal particle surfaces, bridging them to form larger flocs, thereby enhancing the capture of colloids and suspended substances in leachate and improving coagulation performance.

Experimental observations revealed that, without PAM addition, flocs produced by these three coagulants were small, precipitation was ineffective, and the supernatant volume was minimal. As the PAM dosage increased, floc size visibly improved, particularly at 10 mg/L, where flocs compacted, sedimentation accelerated, and the supernatant volume reached 80% of the original leachate volume. Overall, the optimal PAM dosage was determined to be 6 mg/L (Fig. S2c). When PFS was used as a coagulant, its interaction with PAM differed significantly from that of the other three coagulants. COD removal rates fluctuated irrespective of PAM dosage. Observations revealed that flocs generated by PFS without PAM addition were large and settled quickly, producing a larger supernatant volume. However, as the PAM dosage increased, flocs became viscous and

filamentous, with turbidity significantly reduced. Thus, when using PFS, the selection of PAM dosage should primarily focus on turbidity reduction.

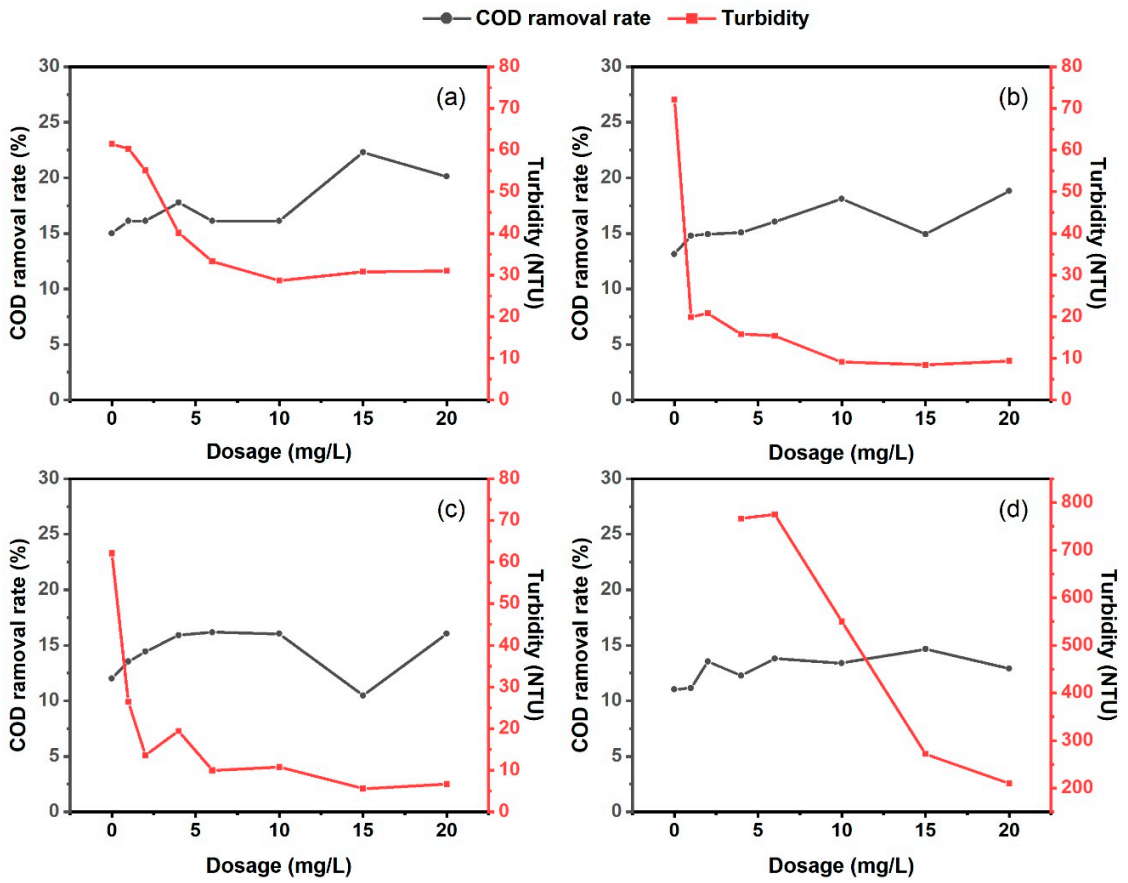


Figure S2. Effect of flocculant dosage on COD and turbidity in leachate (a PSAF; b PAFC; c PAC; d PFS)

The effect of pH conditions

Figure S3 illustrates the influence of initial leachate pH on coagulation performance. When PSAF was used as the coagulant, the COD removal rate remained relatively stable, indicating its effectiveness across a broad pH range. However, turbidity initially increased before decreasing, with the lowest turbidity observed under weakly alkaline conditions. Experimental observations revealed that, in the absence of a coagulant, leachate underwent precipitation under alkaline conditions. As pH increased, more precipitate formed, reducing turbidity and shifting the leachate color from soil brown to a darker shade. During coagulation, increasing pH slowed the precipitation rate, deteriorating the precipitation effect and reducing the supernatant volume. During PSAF coagulation, turbidity reduction observed with increasing pH likely occurred prior to the coagulation process. Although pH had minimal impact on COD, the alkaline environment influenced precipitation characteristics, including floc size and compactness. Based on comprehensive analysis, the optimal pH for PSAF in this study was determined to be 4.

As shown in Figure S3b, PAC exhibited an increasing COD removal rate in a weakly acidic environment (pH 4–7). In contrast, in a weakly alkaline environment (pH 7–9), COD removal rates exhibited fluctuations, suggesting that PAC performed effectively in neutral to weakly alkaline conditions. When PAFC was used as the coagulant, the COD removal rate initially decreased and then increased, peaking at pH 9. This observation indicates that PAFC was effective in acidic and alkaline environments.

In contrast, PFS demonstrated a consistent decline in COD removal rates as pH increased (Figure S3d). Experimental observations revealed that as pH increased, the leachate color deepened, both before and after PFS coagulation. Under weakly alkaline

conditions, the leachate after coagulation lacked clear separation between supernatant and precipitation. Thus, PFS demonstrated the narrowest effective pH range, making it suitable primarily for acidic leachate conditions.

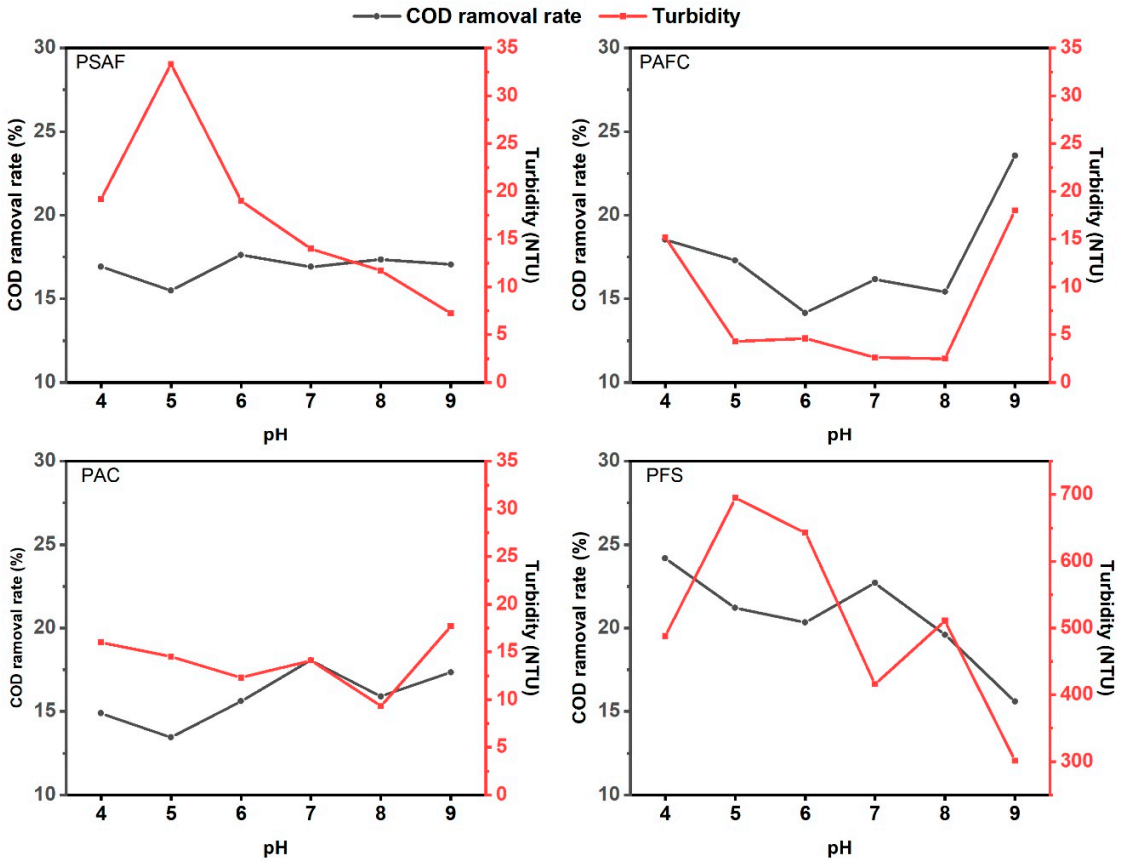


Figure S3. Influence of initial pH of leachate on coagulation effect (a PSAF; b PAFC; c PAC; d PFS)

Experimental materials

The leachate used in this study was collected from a waste transfer station located in Lingang, Shanghai. Immediately after collection, the leachate was refrigerated at 4℃ for 24 hours, followed by filtration through stainless steel meshes with sizes of 6 mesh, 18 mesh, 40 mesh, and 100 mesh. The filtered leachate was then sealed and stored in a dark refrigerator at 4℃ until further use. The basic physicochemical properties of the leachate are presented in Table S1.

Table S1 Characteristics of fresh leachate

Index	**unit**	**concentration**
COD	mg/L	27900
TOC	mg/L	9590
pH	mg/L	4.43
Total nitrogen	mg/L	1060
Ammonia nitrogen	mg/L	423
Nitrate nitrogen	mg/L	8.39

Optimal coagulation pretreatment condition

Based on experimental observations, the effects of four coagulants on COD removal and turbidity reduction in leachate were comprehensively analyzed. The optimal coagulation conditions for each coagulant mixture were identified and are summarized in Table S2.

Table S2 Optimum reaction conditions of four coagulants

	Coagulant dosage (g/L)	Dosage of PAM (mg/L)	Initial pH
PSAF	5	15	-
PAFC	4	10	9
PAC	4	6	7
PFS	4	15	-

Among them, PSAF, PAFC, PAC and PFS are respectively polyferric aluminum silicate, polyferric aluminum chloride, polyaluminum chloride and polyferric sulfate, and "-" means that the pH of the leachate is not changed, as their optimal coagulation performance was achieved under the original pH conditions of the leachate. Preliminary tests showed that pH adjustment did not significantly improve removal efficiency for these two coagulants and might even reduce floc stability. Therefore, the natural pH of the leachate was maintained during their application.

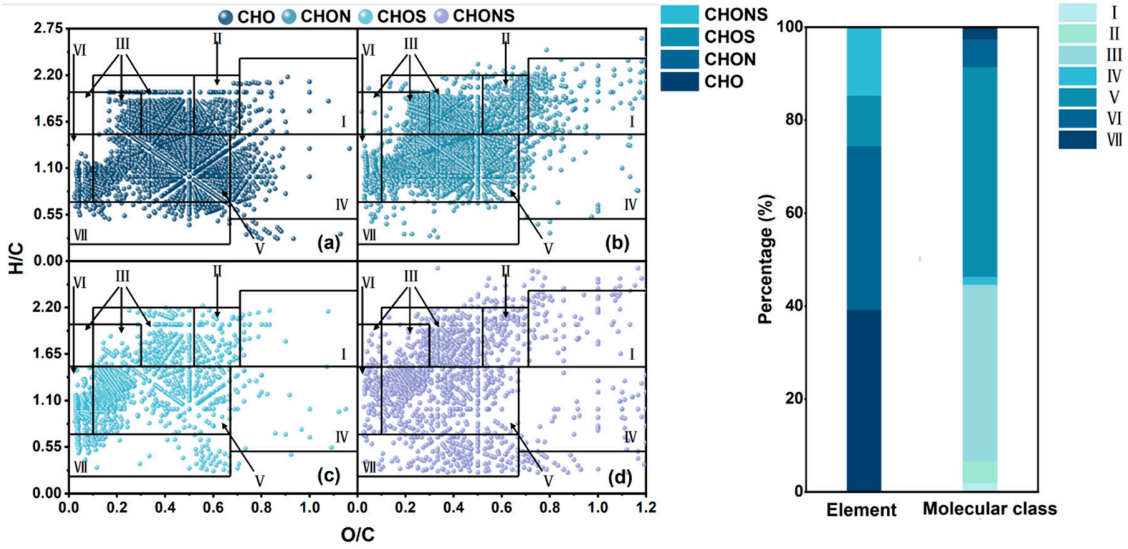


Figure S4. Van Krevelen diagrams representing the four elemental compositions of organic matter in fresh leachate are shown: (a) CHO, (b) CHON, (c) CHOS, and (d) CHONS.

Table S3. Classification and description of four zones in NOSC *vs.* (DBE-O)/C plot.

Classification			
Zone	NOSC	(DBE-O)/C	Description
Zone 1	> 0	≥ 0	Oxidized and unsaturated property
Zone 2	≤ 0	> 0	Reduced and unsaturated property
Zone 3	< 0	≤ 0	Reduced and saturated property
Zone 4	≥ 0	< 0	Oxidized and saturated property
Neutral	= 0	= 0	Neutral state
Definition			
NOSC	A higher (positive) NOSC value indicates a greater oxidation state, a negative NOSC value indicates a reduced state		
(DBE-O)/C	A higher (DBE-O)/C value (positive) indicates that the DOM were more unsaturated, while a more negative value indicates that the DOM are more saturated.		